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<b>UTILITY PATENT APPLICATION TRANSMITTAL</b> (Only for new nonprovisional applications under 37 C.F.R. § 1.53(b))	Attorney Docket No.	HRL033B
	First Inventor or Application Identifier	Shin-Tson Wu
	Title	Polar Tolane Liquid Crystals
	Express Mail Label No.	EK030159612US

<b>APPLICATION ELEMENTS</b> See MPEP chapter 600 concerning utility patent application contents.	<b>ADDRESS TO:</b> Assistant Commissioner for Patents Box Patent Application Washington, DC 20231
1. <input checked="" type="checkbox"/> * Fee Transmittal Form (e.g., PTO/SB/17) (Submit an original and a duplicate for fee processing) 2. <input checked="" type="checkbox"/> Specification [Total Pages 37] (preferred arrangement set forth below) - Descriptive title of the Invention - Cross References to Related Applications - Statement Regarding Fed sponsored R & D - Reference to Microfiche Appendix - Background of the Invention - Brief Summary of the Invention - Brief Description of the Drawings (if filed) - Detailed Description - Claim(s) - Abstract of the Disclosure 3. <input type="checkbox"/> Drawing(s) (35 U.S.C. 113) [Total Sheets ] 4. Oath or Declaration [Total Pages 4] a. <input checked="" type="checkbox"/> Newly executed (original or copy) b. <input type="checkbox"/> Copy from a prior application (37 C.F.R. § 1.63(d)) (for continuation/divisional with Box 16 completed) i. <input type="checkbox"/> <b>DELETION OF INVENTOR(S)</b> Signed statement attached deleting inventor(s) named in the prior application, see 37 C.F.R. §§ 1.63(d)(2) and 1.33(b).	5. <input type="checkbox"/> Microfiche Computer Program (Appendix) 6. Nucleotide and/or Amino Acid Sequence Submission (if applicable, all necessary) a. <input type="checkbox"/> Computer Readable Copy b. <input type="checkbox"/> Paper Copy (identical to computer copy) c. <input type="checkbox"/> Statement verifying identity of above copies <b>ACCOMPANYING APPLICATION PARTS</b> 7. <input type="checkbox"/> Assignment Papers (cover sheet & document(s)) 8. <input type="checkbox"/> 37 C.F.R. § 3.73(b) Statement of Power of Attorney (when there is an assignee) 9. <input type="checkbox"/> English Translation Document (if applicable) 10. <input type="checkbox"/> Information Disclosure Statement (IDS)/PTO-1449 [Copies of IDS Citations] 11. <input type="checkbox"/> Preliminary Amendment 12. <input checked="" type="checkbox"/> Return Receipt Postcard (MPEP 503) (Should be specifically itemized) 13. <input type="checkbox"/> * Small Entity Statement filed in prior application, Status still proper and desired (PTO/SB/09-12) 14. <input type="checkbox"/> Certified Copy of Priority Document(s) (if foreign priority is claimed) 15. <input type="checkbox"/> Other:

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☐ Customer Number or Bar Code Label \_\_\_\_\_ or ☒ Correspondence address below  
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Name	Cary Tope-McKay				
Address	23852 Pacific Coast Highway #311				
City	Malibu	State	CA	Zip Code	90265
Country	USA	Telephone	(310) 589-8158	Fax	(310) 943-2736

Name (Print/Type)	Cary Tope-McKay	Registration No. (Attorney/Agent)	41,350
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See 37 C.F.R. §§ 1.27 and 1.28.

TOTAL AMOUNT OF PAYMENT (\$996

## Complete if Known

Application Number  
Filing Date  
First Named Inventor Shin-Tson Wu  
Examiner Name  
Group / Art Unit  
Attorney Docket No. HRL033B

PTO  
U.S.  
09/05/00



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101 690	201 345	Utility filing fee	\$690
106 310	206 155	Design filing fee	
107 480	207 240	Plant filing fee	
108 690	208 345	Reissue filing fee	
114 150	214 75	Provisional filing fee	

SUBTOTAL (1) (\$690

#### 2. EXTRA CLAIM FEES

	Extra Claims	Fee from below	Fee Paid
Total Claims 24	-20** = 4	18	72
Independent Claims 6	-3** = 3	78	234
Multiple Dependent		0	0

\*\*or number previously paid, if greater; For Reissues, see below

Large Entity Fee Code (\$)	Small Entity Fee Code (\$)	Fee Description
103 18	203 9	Claims in excess of 20
102 78	202 39	Independent claims in excess of 3
104 260	204 130	Multiple dependent claim, if not paid
109 78	209 39	** Reissue independent claims over original patent
110 18	210 9	** Reissue claims in excess of 20 and over original patent

SUBTOTAL (2) (\$306

### FEE CALCULATION (continued)

#### 3. ADDITIONAL FEES

Large Entity Fee Code (\$)	Small Entity Fee Code (\$)	Fee Description	Fee Paid
105 130	205 65	Surcharge - late filing fee or oath	
127 50	227 25	Surcharge - late provisional filing fee or cover sheet.	
139 130	139 130	Non-English specification	
147 2,520	147 2,520	For filing a request for reexamination	
112 920*	112 920*	Requesting publication of SIR prior to Examiner action	
113 1,840*	113 1,840*	Requesting publication of SIR after Examiner action	
115 110	215 55	Extension for reply within first month	
116 380	216 190	Extension for reply within second month	
117 870	217 435	Extension for reply within third month	
118 1,360	218 680	Extension for reply within fourth month	
128 1,850	228 925	Extension for reply within fifth month	
119 300	219 150	Notice of Appeal	
120 300	220 150	Filing a brief in support of an appeal	
121 260	221 130	Request for oral hearing	
138 1,510	138 1,510	Petition to institute a public use proceeding	
140 110	240 55	Petition to revive - unavoidable	
141 1,210	241 605	Petition to revive - unintentional	
142 1,210	242 605	Utility issue fee (or reissue)	
143 430	243 215	Design issue fee	
144 580	244 290	Plant issue fee	
122 130	122 130	Petitions to the Commissioner	
123 50	123 50	Petitions related to provisional applications	
126 240	126 240	Submission of Information Disclosure Stmt	
581 40	581 40	Recording each patent assignment per property (times number of properties)	
146 690	246 345	Filing a submission after final rejection (37 CFR § 1.129(a))	
149 690	249 345	For each additional invention to be examined (37 CFR § 1.129(b))	

Other fee (specify) \_\_\_\_\_

Other fee (specify) \_\_\_\_\_

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SUBTOTAL (3) (\$0

### SUBMITTED BY

Name (Print/Type) Cary Tope-McKay  
Registration No. (Attorney/Agent) 41,350  
Telephone (310) 589-8158  
Signature  
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# **POLAR TOLANE LIQUID CRYSTALS**

## **FIELD OF THE INVENTION**

The present invention relates to liquid crystal materials and eutectic mixtures thereof. In particular, the invention is directed to a new class of polar asymmetric tolane derivatives  
5 and eutectic liquid crystal mixtures containing the derivatives with infrared and microwave applications.

## **BACKGROUND OF THE INVENTION:**

Liquid crystals that have high birefringence, low viscosity, and low threshold voltage,  
10 and are capable of maintaining a nematic phase over a broad temperature range are desirable in electro-optic phase and amplitude modulation applications. Such applications include infrared light valves, polymer-dispersed liquid crystals and cholesteric displays.

High birefringence, or optical anisotropy ( $\Delta n$ ), of the liquid crystal composition improves the efficiency of light modulation, and low rotational viscosity serves to shorten the  
15 response times. Low threshold voltage, which is inversely related to the dielectric anisotropy of the liquid crystal material, simplifies the electronics that drive the application.

Polar liquid crystal compounds, particularly fluorinated species, have been investigated  
20 as possible candidates for such electro-optics applications. In general, fluorinated liquid crystal compounds have desirably low rotational viscosity, excellent photostability and

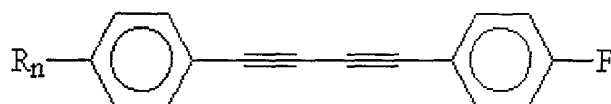
high resistivity. However, they also tend to exhibit modest dielectric anisotropy and, accordingly, high threshold voltage.

Increasing the asymmetry and, accordingly, the polarity of such compounds has been

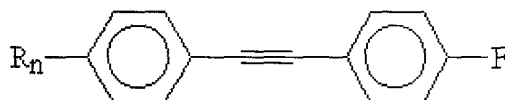
5 proposed as a means to improve the dielectric anisotropy of these compounds. Wu, et al.

[Opt. Eng. 32, 1792-7 (1993)], for example, reported polar fluorinated

diphenyldiacetylenes and tolanes as shown in Structures I and II below, respectively



(Structure I)

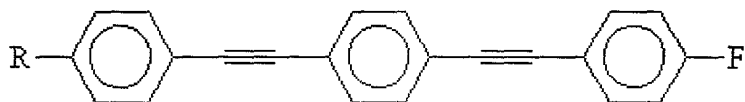


(Structure II)

10 These compounds exhibit high birefringence, high resistivity, low viscosity, and comparatively low threshold voltage. However, while in some cases these compounds have reasonably low melting temperatures and heat fusion enthalpies, as is typical of

15 compounds with high dipole moments, the melting temperature is too high and the nematic phase is too narrow.

The fluorination of polytolane liquid crystal compounds has also been investigated. In general, the highly conjugated polytolanes have desirably high birefringence. Fluorinated bis-tolanes such as shown in structure III, have been reported.

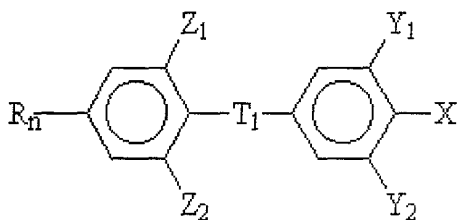


5 (Structure III)

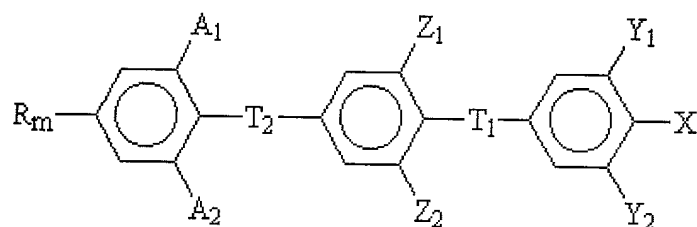
While this type of compound has good dielectric anisotropy, utility is limited, once again, due to high melting point and narrow nematic range.

### SUMMARY OF THE INVENTION

- 10 A new class of liquid crystal compounds is provided which provides improved melting points and broader nematic ranges compared to previously reported fluorinated liquid crystal compounds. Specifically, the present invention provides a class of compounds based on the tolane and bis-tolane structures as shown in Structures IV and V.



15 (Structure IV)

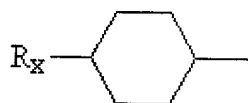


(Structure V)

For the compounds depicted in all of the above structures and described herein, a polar group such as F (fluoro), CN (cyano), OCF<sub>3</sub> (trifluoromethoxy), or NCS (isothiocyanate) is positioned at site X and fluoro groups are positioned at least one of the pairs of sites Y<sub>1</sub> and Y<sub>2</sub>, Z<sub>1</sub> and Z<sub>2</sub>, and for the bis-tolane derivatives, A<sub>1</sub> and A<sub>2</sub>.

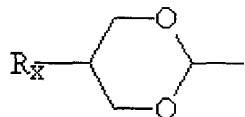
For the tolane derivatives as depicted in structure IV, T<sub>1</sub> is always a triple bond. For the bis-tolane derivatives, T<sub>1</sub> and T<sub>2</sub> are either both triple bonds or one of the two groups is a double bond with and the other remains a triple bond.

In this new class of compounds, R<sub>n</sub> or R<sub>m</sub> may be an alkyl group having the general formula C<sub>n</sub>H<sub>2n+1</sub>, an alkenyl group having the general formula C<sub>n</sub>H<sub>2n-1</sub>, an alkoxy group having the general formula OC<sub>n</sub>H<sub>2n+1</sub>, or an alkenoxy group having the general formula – OC<sub>n</sub>H<sub>2n-1</sub>. Additionally, for the tolane compounds depicted in structure IV, R<sub>n</sub> may be a cyclohexyl substituent as shown in Structure VI



(Structure VI)

or a dioxane substituent as shown in Structure VII.



(Structure VII)

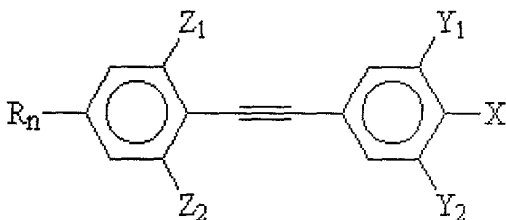
- 5 For both types of substituents,  $R_X$  may be an alkyl group having the general formula  
general formula  $-C_xH_{2x+1}$ , an alkenyl group having the general formula  $-C_xH_{2x-1}$ , an  
alkoxy group having the general formula  $-OC_xH_{2x+1}$ , or an alkenoxy group having the  
general formula  $-OC_xH_{2x-1}$ .

## DETAILED DESCRIPTION OF THE INVENTION

The present invention relates to a new class of liquid crystal materials and eutectic  
mixtures thereof and a method for their synthesis. It is anticipated that the invention may  
be tailored to a variety of other applications. The following description is presented to  
enable one of ordinary skill in the art to make and use the invention and to incorporate it  
15 in the context of particular applications. Various modifications, as well as a variety of  
uses in different applications, will be readily apparent to those skilled in the art, and the  
general principles defined herein may be applied to a wide range of embodiments. Thus,  
the present invention is not intended to be limited to the embodiments presented, but is to  
be accorded the widest scope consistent with the principles and novel features disclosed  
20 herein.

The present invention relates, generally, to fluorinated polar asymmetric liquid crystal compound, a method for their preparation, and their use in eutectic mixtures. In certain embodiments of this invention, tolane compounds are provided of the type depicted in

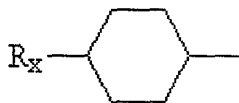
5 Structure VIII:



(Structure VIII)

wherein X is a polar group such as F (fluoro), CN (cyano),  $OCF_3$  (trifluoromethoxy), or NCS (isothiocyanate), and at least one of the pairs  $Y_1$  and  $Y_2$  and  $Z_1$  and  $Z_2$  are fluoro groups. In these embodiments,  $R_n$  may be an alkyl group having the general formula  $C_nH_{2n+1}$ , an alkenyl group having the general formula  $C_nH_{2n-1}$ , an alkoxy group having the general formula  $OC_nH_{2n+1}$ , or an alkenoxy group having the general formula  $OC_nH_{2n-1}$ . For these substituents, the value of n is preferably approximately 2 to 12.

Additionally,  $R_n$  may be a substituent of the type shown in Structure VI,

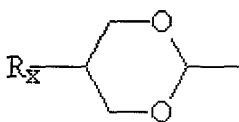


(Structure VI)

wherein  $R_x$  is an alkyl group having the general formula  $C_xH_{2x+1}$ , an alkenyl group having the general formula  $C_xH_{2x-1}$ , an alkoxy group having the general formula



$\text{OC}_x\text{H}_{2x+1}$ , or an alkenoxy group having the general formula  $\text{OC}_x\text{H}_{2x-1}$ . The value of  $x$  is preferably approximately 2 to 12. In yet another alternative,  $\text{R}_n$  may be a substituent of the type shown in Structure VII,

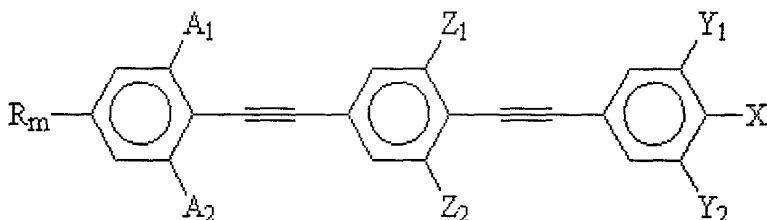


5 (Structure VII)

wherein again  $\text{R}_x$  may be an alkyl group having the general formula  $\text{C}_x\text{H}_{2x+1}$ , an alkenyl group having the general formula  $\text{C}_x\text{H}_{2x-1}$ , an alkoxy group having the general formula  $\text{OC}_x\text{H}_{2x+1}$ , or an alkenoxy group having the general formula  $\text{OC}_x\text{H}_{2x-1}$ . Again, the value of  $x$  is preferably approximately 2 to 12.

10

In other, preferred embodiments of this invention, bis-tolane compounds are provided as shown in structure IX,



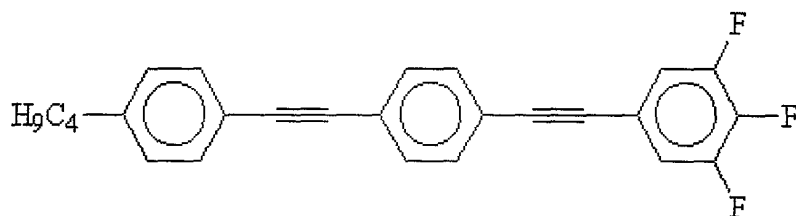
(Structure IX)

- 15 wherein  $X$  is a polar group such as  $F$  (fluoro),  $\text{CN}$  (cyano),  $\text{OCF}_3$  (trifluoromethoxy), or  $\text{NCS}$  (isothiocyanate), and at least one of the pairs  $Y_1$  and  $Y_2$ ,  $Z_1$  and  $Z_2$ , and  $A_1$  and  $A_2$  are fluoro groups. In this embodiment,  $\text{R}_m$  may be an alkyl group having the general formula  $\text{C}_m\text{H}_{2m+1}$ , an alkenyl group having the general formula  $\text{C}_m\text{H}_{2m-1}$ , an alkoxy group

having the general formula  $\text{OC}_m\text{H}_{2m+1}$ , or an alkenoxy group having the general formula  $\text{OC}_m\text{H}_{2m-1}$ . The value of  $m$  is preferably approximately 2 to 12.

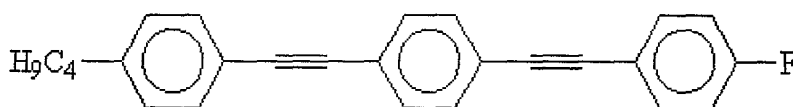
One example of this embodiment, wherein  $R_m$  is an alkyl group with  $m=4$ , is depicted

5 for clarity in structure X and will henceforth be referred to as PTPTP-4FFF.



(Structure X)

This trifluoro bis-tolane compound has a nematic range of 115 to 188.1 °C and a heat fusion enthalpy of 3.85 kcal/mol. In comparison, the monofluoro compound, henceforth  
10 referred to as PTPTP-4F, and depicted in structure XI,



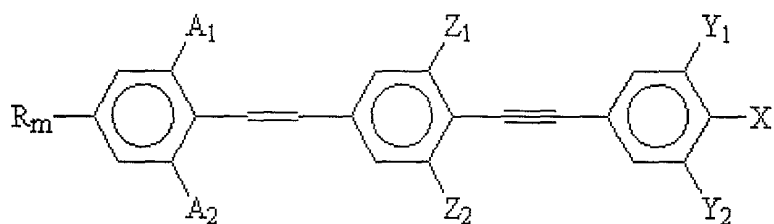
(Structure XI)

has a nematic range of 173.1 to 217.2 °C and a heat fusion enthalpy of 8.47 kcal/mol.

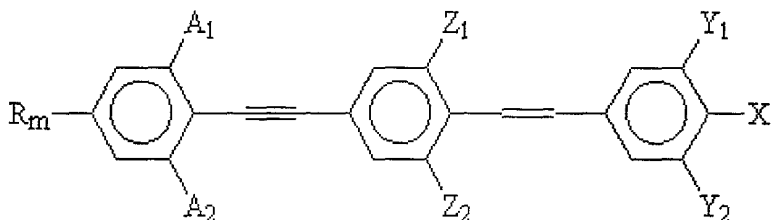
The addition of the fluoro groups meta to the tolane triple bond serves to reduce both the  
15 melting and clearing points of the compound and accordingly reduce the heat fusion enthalpy. Further, in contrast to PTPTP-4F, the reduced melting point of the PTPTP-4FFF species renders it useful in eutectic mixtures.

In yet other embodiments, modified bis-tolane structures of the types depicted in

Structures XII and XIII are provided:



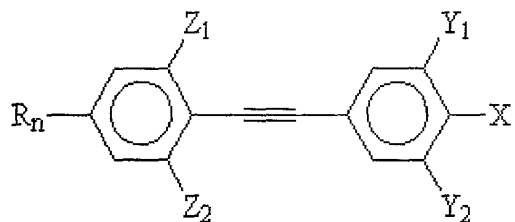
(Structure XII)



(Structure XIII)

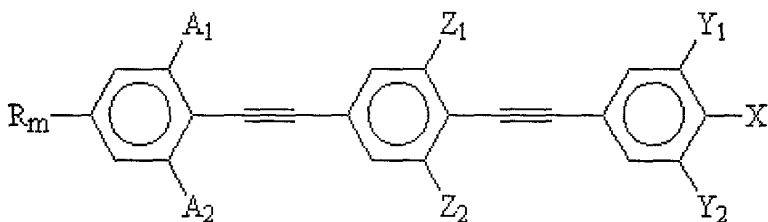
wherein X is a polar group such as F (fluoro), CN (cyano), OCF<sub>3</sub> (trifluoromethoxy), or NCS (isothiocyanate), and either the pair Y<sub>1</sub> and Y<sub>2</sub>, the pair Z<sub>1</sub> and Z<sub>2</sub>, or the pair A<sub>1</sub> and A<sub>2</sub> are fluoro groups and the alternate pairs are hydrogens. In this embodiment, R<sub>m</sub> may be an alkyl group having the general formula C<sub>m</sub>H<sub>2m+1</sub>, an alkenyl group having the general formula C<sub>m</sub>H<sub>2m-1</sub>, an alkoxy group having the general formula OC<sub>m</sub>H<sub>2m+1</sub>, or an alkenoxy group having the general formula OC<sub>m</sub>H<sub>2m-1</sub>. The value of m is preferably approximately 2 to 12.

For preferred embodiments of the tolans provided by the present invention as shown in structure VIII,

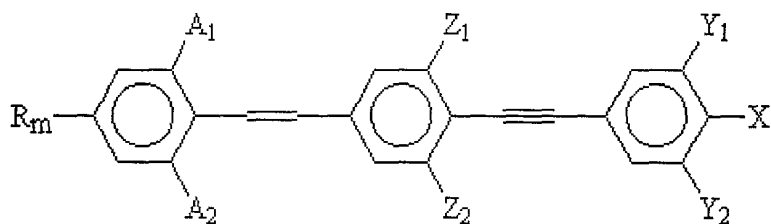


(Structure VIII)

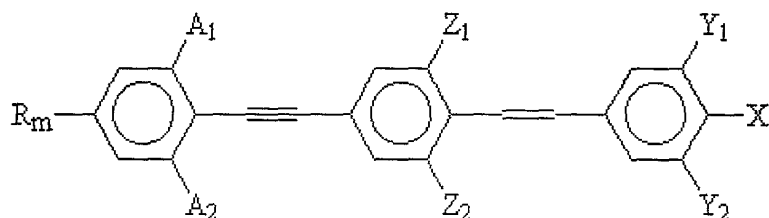
the  $R_n$  substituent is an alkenyl group having the general formula  $C_nH_{2n-1}$ , and more preferably an alkenyl group with the double bond in the second position and having the  
 5 general formula  $C_xH_{2x+1}CH=CH-CH_2-$ . Similarly for the bis-tolanes and modified bis-tolane derivatives shown structures IX, XII, and XIII,



(Structure IX)



10 (Structure XII)

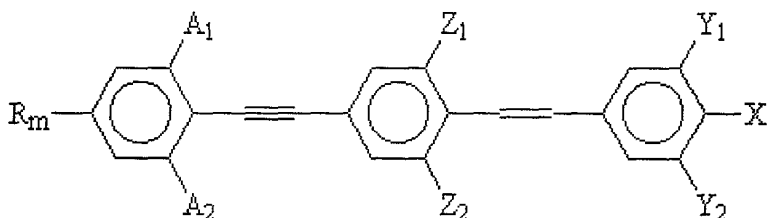


(Structure XIII)

$R_m$  is preferably an alkenyl group with the general formula  $C_mH_{2m-1}$ , and more preferably an alkenyl with the double bond in the second position and having the general formula

- 5  $C_xH_{2x+1}CH=CH-CH_2-$ . Furthermore, for the tolanes, bis-tolanes, and modified bis-tolanes provided herein, fluoro substitution at the X,  $Y_1$ , and  $Y_2$  positions is preferred.

Generally, the compounds provided in the present invention have large dipole moments that contribute to desirably high dielectric anisotropy. However, the positioning of the  
 10 additional fluoro groups helps to reduce the high melting points characteristic of polar tolane and bis-tolane compounds. Modified bis-tolane structure XIII is shown here for reference.



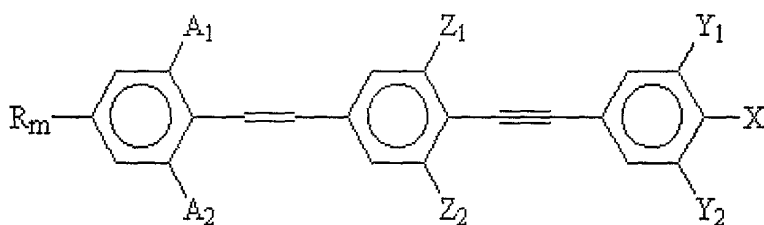
(Structure XIII)

- 15 However, the principles presented herein apply to all embodiments of the present invention. It should be apparent to one skilled in the art that placing fluoro groups in at

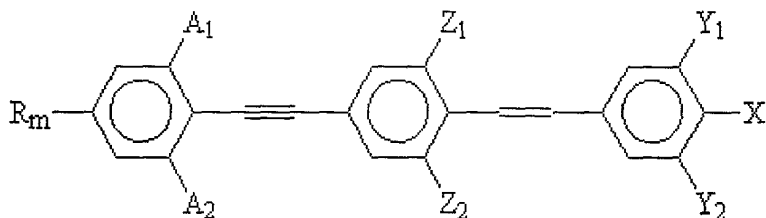
least one pair of the  $Y_1$ ,  $Y_2$ , or  $Z_1$ ,  $Z_2$ , or  $A_1$ ,  $A_2$  paired positions in the above structure not only contributes to the overall dipole of the molecule but also serves to increase the effective molecular distance between molecules in the bulk phase. Increasing the intermolecular distance results in a decrease in melting point and heat fusion enthalpy.

5

For the modified bis-tolane embodiments of the present invention, reduction of one of the bis-tolane triple bonds to a double bond as shown in structures XII and XIII



(Structure XII)

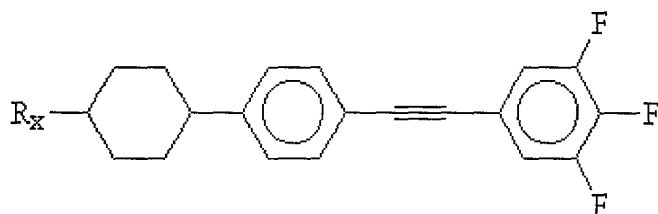


(Structure XIII)

introduces a bend into the molecular structure. This bend, like the pairs of fluoro substituents, serves to increase the intermolecular distance in the bulk phase. Thus, the reduction of one of the triple bonds in the bis-tolane structure results in a lower melting point and heat fusion enthalpy for the compound.

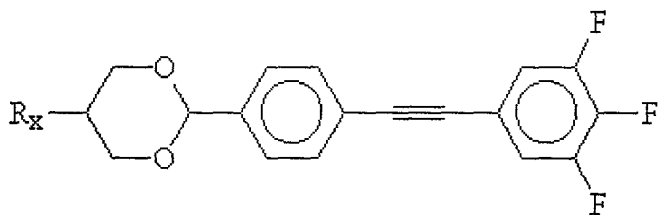
15

In additional embodiments of the present invention, the compounds provided herein are utilized as components of eutectic mixtures. In general, compounds such as those depicted in structure XIV



5 (Structure IV)

are particularly effective due to low melting point and wide nematic range. Compounds such as PTPTP-4FFF and the class of compounds depicted in Structure XV,



(Structure XV)

10 while having relatively high melting points and limited nematic phases, are nonetheless useful as dopants for increasing the dielectric anisotropy of eutectic mixtures.

Compounds of the type shown in structure XV, in particular, have very high dipole moments and accordingly large dielectric anisotropy values.

15 General procedures for the preparation of embodiments of this invention are provided in Schemes 1 and 2. Scheme 1 outlines particularly the synthesis of the tolane compounds

(4), and Scheme 2 outlines particularly the bis-tolane variants (11). A detailed description of a synthesis example compound PTPTP-4FFF according to Scheme 2 is provided below. It will be readily apparent to one skilled in the art that the synthesis for PTPTP-4FFF described herein can be readily adapted, as outlined in Schemes 1 and 2, to produce other variants of the present invention. Appropriate selection of reactants, such as substituted benzenes, as outlined in the Schemes below will provide the desired products according to the present invention.

To a mixture of (trimethylsilyl)acetylene **1a** (5.0 g, 51 mmol) and 1-butyl-4-iodobenzene **5** (10.6 g, 46 mmol) was added the catalyst system of Pd(PPh<sub>3</sub>)Cl<sub>2</sub> (800 mg, 1.2 mmol) and CuI (220 mg, 1.2 mmol) followed by anhydrous tetrahydrofuran (50 mL). The mixture was kept under an Ar atmosphere and cooled with an ice-bath. Triethylamine (10 mL, 72 mmol) was added to the mixture through a syringe over 5 minutes and the orange solution was stirred for 1.5 hours while the ice-bath warmed up. The resulting dark solution was diluted with pentane and washed with 10% HCl, saturated NaHCO<sub>3</sub>, and brine, successively. The organic layer was dried with Na<sub>2</sub>SO<sub>4</sub>, concentrated and passed through a plug of silica gel. Evaporation of solvent gave the trimethylsilyl acetylene derivative **6** as a clear oil (9.2 g, 100%).

To **6** (m = 4) (9.2 g, 46 mmol) was added a solution of NaOH (4 g, 100 mmol) in a mixed solvent of H<sub>2</sub>O (8 mL), THF (8 mL) and MeOH (8 mL). The mixture was stirred at room temperature for 17 hours. It was then diluted with hexanes and washed with 10% HCl, saturated NaHCO<sub>3</sub> and brine, successively. The organic layer was dried with Na<sub>2</sub>SO<sub>4</sub>,



concentrated and passed through a plug of silica gel. Evaporation of solvent gave the unsubstituted acetylene compound **7** as a clear oil (5.5 g, 93%).

To an ice-bath-cooled mixture of **7** ( $m = 4$ ) (1.88 g, 11.9 mmol), 1-bromo-4-iodobenzene  
5 **7a** (3.06 g, 10.8 mmol),  $\text{Pd}(\text{PPh}_3)\text{Cl}_2$  (209 mg, 0.3 mmol) and  $\text{CuI}$  (57 mg, 0.3 mmol)  
were added tetrahydrofuran (12 mL) followed by triethylamine (5 mL). The mixture  
was stirred for 24 hours at room temperature. The precipitates were removed by vacuum  
filtration and the filtrate was diluted with pentane. The organic layer was washed with  
10%  $\text{HCl}$ , saturated  $\text{NaHCO}_3$  and brine, successively. After drying with  $\text{MgSO}_4$ ,  
10 concentration and filtration through silica gel, the brominated tolane **8** was obtained as a  
yellow solid (3.22 g, 95%).

To a solution of **8** ( $m = 4$ ) (743 mg, 2.4 mmol),  $\text{Pd}(\text{PPh}_3)\text{Cl}_2$  (28 mg, 0.01 mmol),  $\text{CuI}$  (5  
mg, 0.01 mmol) and  $\text{PPh}_3$  (10 mg, 0.01 mmol) in triethylamine (10 mL) was added  
15 (trimethylsilyl)acetylene (500  $\mu\text{L}$ , 3.6 mmol). The solution was refluxed under Ar for 1.5  
hours and then cooled to room temperature. Hexane extracts of the mixture were washed  
with 10%  $\text{HCl}$ , saturated  $\text{NaHCO}_3$ , and brine, successively. The solution was dried with  
 $\text{Na}_2\text{SO}_4$  and concentrated. The crude oil was subject to column chromatography to give a  
yellowish solid (604 mg, 84%) as the pure trimethylsilyl acetylene derivative **9**.

20 To a solution of **9** ( $m = 4$ ) (430 mg, 1.3 mmol) in the mixed solvent of tetrahydrofuran (6  
mL) and ethanol (6 mL) was added 1 g of Dowex® 550A  $\text{OH}^-$  anion-exchange resin.

The mixture was stirred at room temperature for 18 hours. After the resin was filtered, the solution was concentrated and filtered through silica gel (hexanes as eluent). Evaporation of solvent gave the unsubstituted acetylene derivative **10** as a yellow solid (350 mg, 100%).

5

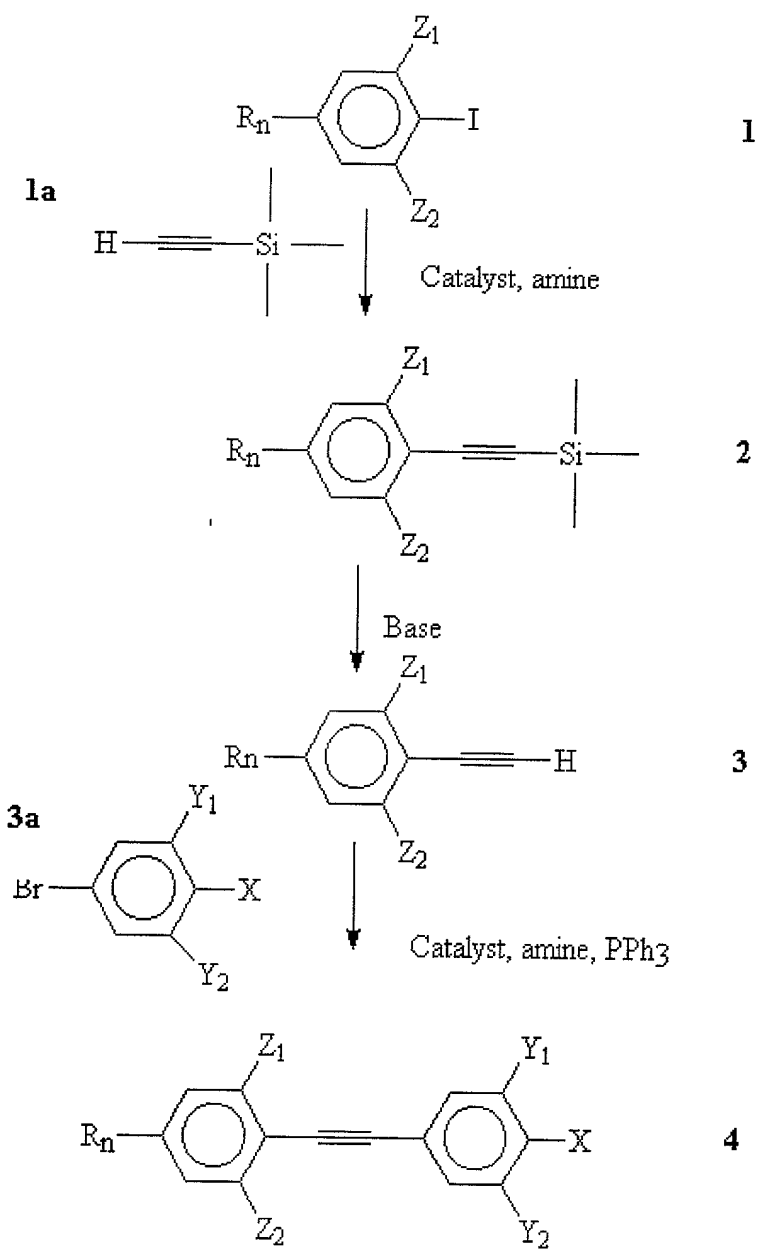
To a mixture of **10** ( $n = 4$ ) (182 mg, 1 mmol) and 1-bromo-3,4,5-trifluorobenzene **10a** (211 mg, 1 mmol) was added Pd(PPh<sub>3</sub>)<sub>4</sub> (12 mg, 0.01 mmol), CuI (2 mg, 0.01 mmol) and PPh<sub>3</sub> (4 mg, 0.01 mmol) followed by triethylamine (5 mL). The mixture was refluxed under Ar for 2 hours and cooled down to room temperature. It was then diluted with hexanes, filtered through silica gel and concentrated. Column chromatography of the crude oil gave the pure bis-tolane product **11** as a clear oil (80%).

10

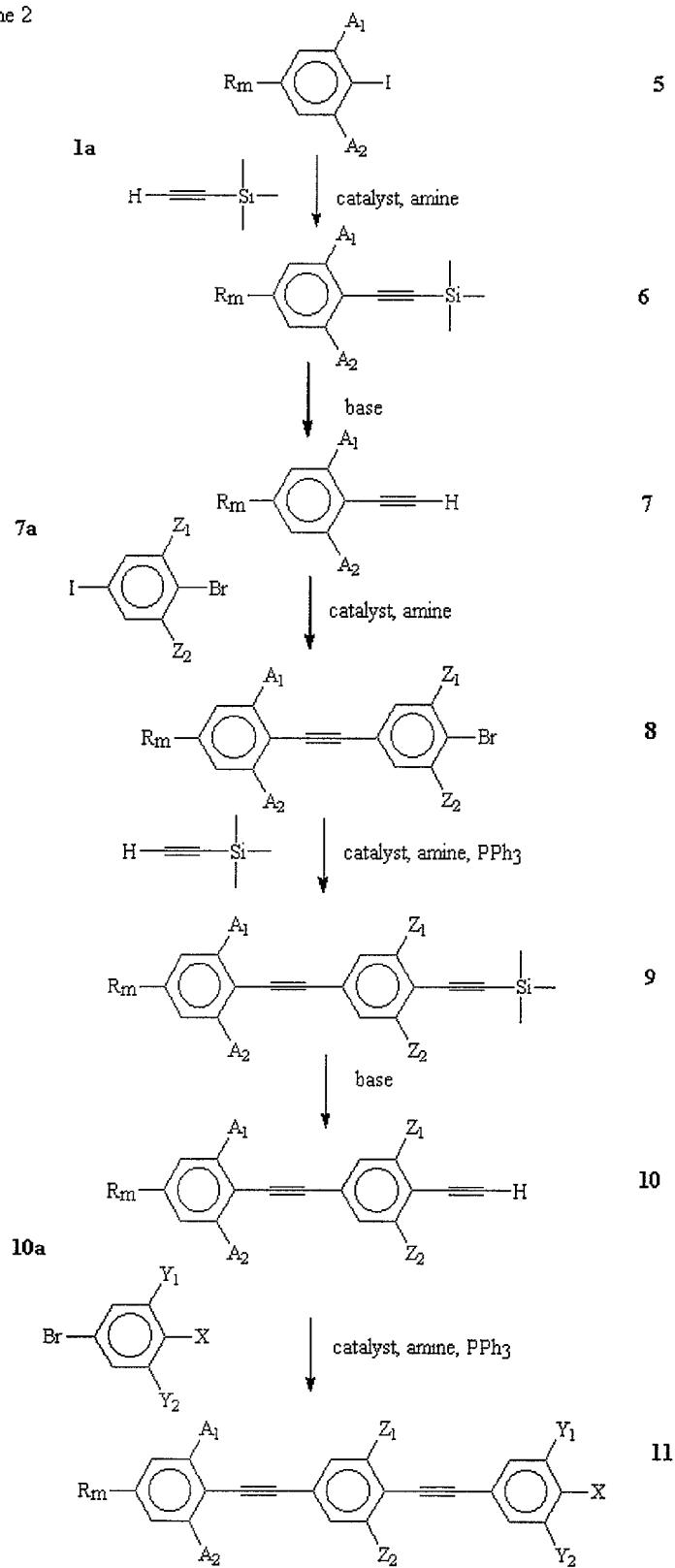
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Scheme 1



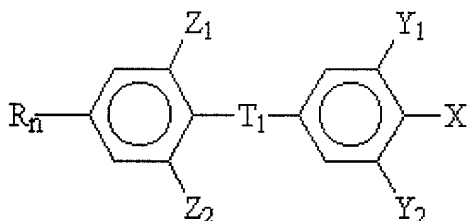
Scheme 2



## CLAIMS

What is claimed is:

1. Liquid crystal compounds having the general structure:



5 (Structure IV)

wherein X is selected from the group consisting of F (fluoro), CN (cyano),

$OCF_3$ (trifluoromethoxy), and NSC(isothiocyanate);

$T_1$  is a triple bond;

$Y_1$  and  $Y_2$  are a pair of substituents selected from the group consisting of H and F, and  $Y_1$

10 =  $Y_2$ ;

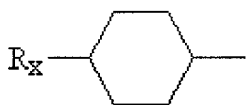
$Z_1$  and  $Z_2$  are a pair of substituents selected from the group consisting of H and F, and  $Z_1$

=  $Z_2$ ; and,

at least one of the pairs  $Y_1$  and  $Y_2$  and  $Z_1$  and  $Z_2$  are substituted with F;

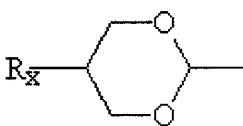
$R_n$  is selected from the group consisting of an alkyl group having the general formula

15  $C_nH_{2n+1}$ , an alkenyl group having the general formula  $C_nH_{2n-1}$ , an alkoxy group having the general formula  $OC_nH_{2n+1}$ , an alkenoxy group having the general formula  $OC_nH_{2n-1}$ , a group of the general structure



(Structure VI)

and a group of the general structure



5 (Structure VII)

wherein  $R_X$  for both structures is selected from a group consisting of an alkyl group having the general formula  $C_xH_{2x+1}$ , an alkenyl group having the general formula  $C_xH_{2x-1}$ , an alkoxy group having the general formula  $OC_xH_{2x+1}$ , and an alkenoxy group having the general formula  $OC_xH_{2x-1}$ .

10

2. A liquid crystal compound as set forth in claim 1, wherein X is substituted with F; and,  $Y_1$  and  $Y_2$  are substituted with F and  $Z_1$  and  $Z_2$  are H groups.

15

3. A liquid crystal compound as set forth in claim 1, wherein  $R_n$  is selected from a group consisting of an alkyl group having the general formula  $C_nH_{2n+1}$ , an alkenyl group having the general formula  $C_nH_{2n-1}$ , an alkoxy group having the general formula  $OC_nH_{2n+1}$ , and an alkenoxy group having the general formula  $OC_nH_{2n-1}$  where n is approximately 2 to 12.

4. A liquid crystal compound as set forth in claim 1, wherein  $R_x$  is selected from a group consisting of an alkyl group having the general formula  $C_xH_{2x+1}$ , an alkenyl group having the general formula  $C_xH_{2x-1}$ , an alkoxy group having the general formula  $OC_xH_{2x+1}$ , and an alkenoxy group having the general formula  $OC_xH_{2x-1}$  where  $x$  is approximately 2 to

5 12.

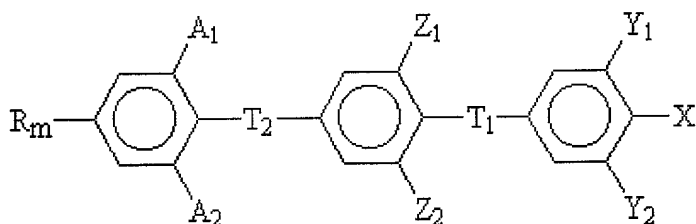
5. A liquid crystal compound as set forth in claim 1, wherein  $R_n$  is an alkenyl group having the general formula  $C_nH_{2n-1}$ .

10 6. A liquid crystal compound as set forth in claim 1, wherein  $R_n$  is an alkenyl group having the general formula  $C_nH_{2n-1}$  where  $n$  ranges approximately from 2 to 12.

7. A liquid crystal compound as set forth in claim 1, wherein  $R_n$  is an alkenyl group having the general formula  $C_xH_{2x-1}CH=CH-(CH_2)-$ .

15

8. Liquid crystal compounds having the general structure



(Structure V)

wherein X is selected from the group consisting of F (fluoro), CN (cyano),

OCF<sub>3</sub>(trifluoromethoxy), and NSC(isothiocyanate);

T<sub>1</sub> is selected from the group consisting of a triple and a double covalent bond between

5 two carbons;

T<sub>2</sub> is selected from the group consisting of a triple and a double covalent bond between

two carbons; and,

T<sub>1</sub> is not equal to T<sub>2</sub> when T<sub>1</sub> or T<sub>2</sub> is a double bond;

Y<sub>1</sub> and Y<sub>2</sub> are a pair of substituents selected from the group consisting of H and F and Y<sub>1</sub>

10 = Y<sub>2</sub>;

Z<sub>1</sub> and Z<sub>2</sub> are a pair of substituents selected from the group consisting of H and F and Z<sub>1</sub>

= Z<sub>2</sub>;

A<sub>1</sub> and A<sub>2</sub> are a pair of substituents selected from the group consisting of H and F and A<sub>1</sub>

= A<sub>2</sub>;

15 at least one of the pairs Y<sub>1</sub> and Y<sub>2</sub>, Z<sub>1</sub> and Z<sub>2</sub>, and A<sub>1</sub> and A<sub>2</sub> is substituted with F; and,

R<sub>m</sub> is selected from the group consisting of selected from a group consisting of an alkyl

group having the general formula C<sub>m</sub>H<sub>2m+1</sub>, an alkenyl group having the general formula

C<sub>m</sub>H<sub>2m-1</sub>, an alkoxy group having the general formula OC<sub>m</sub>H<sub>2m+1</sub>, and an alkenoxy group

having the general formula OC<sub>m</sub>H<sub>2m-1</sub>.

20

9. A liquid crystal compound as set forth in claim 8, wherein X is a substituted with F;



$Y_1$  and  $Y_2$  are substituted with F; and,

$Z_1$  and  $Z_2$  and  $A_1$  and  $A_2$  are H groups.

10. A liquid crystal compound as set forth in claim 8, wherein  $T_1$  and  $T_2$  are triple bonds  
5 between two carbons.

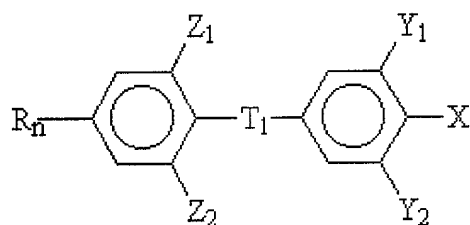
11. A liquid crystal compound as set forth in claim 8, wherein  $R_m$  is selected from a  
group consisting of an alkyl group having the general formula  $C_mH_{2m+1}$ , an alkenyl group  
having the general formula  $C_mH_{2m-1}$ , an alkoxy group having the general formula  
10  $OC_mH_{2m+1}$ , and an alkenoxy group having the general formula  $OC_mH_{2m-1}$  where m is  
approximately 2 to 12.

12. A liquid crystal compound as set forth in claim 8, wherein  $R_m$  is an alkenyl group  
having the general formula  $C_mH_{2m-1}$ .  
15

13. A liquid crystal compound as set forth in claim 8, wherein  $R_m$  is an alkenyl group  
having the general formula  $C_mH_{2m-1}$  where m ranges approximately from 2 to 12.

14. A liquid crystal compound as set forth in claim 8, wherein  $R_m$  is an alkenyl group  
20 having the general formula  $C_mH_{2m-1}CH=CH-(CH_2)-$ .

15. A eutectic mixture of liquid crystal compounds comprising at least two liquid crystal compounds, including at least one compound having the general structure



(Structure IV)

- 5 wherein X is selected from the group consisting of F (fluoro), CN (cyano), OCF<sub>3</sub>(trifluoromethoxy), and NSC(isothiocyanate);

$T_1$  is a triple bond;

$Y_1$  and  $Y_2$  are a pair of substituents selected from the group consisting of H and F and  $Y_1 = Y_2$ ;

- 10  $Z_1$  and  $Z_2$  are a pair of substituents selected from the group consisting of H and F and  $Z_1 = Z_2$ ;

at least one of the pairs  $Y_1$  and  $Y_2$  and  $Z_1$  and  $Z_2$  are substituted with F;

$R_n$  is selected from the group consisting of an alkyl group having the general formula

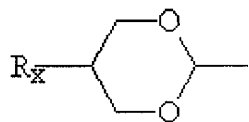
$C_nH_{2n+1}$ , an alkenyl group having the general formula  $C_nH_{2n-1}$ , an alkoxy group having

- 15 the general formula  $OC_nH_{2n+1}$ , an alkenoxy group having the general formula  $OC_nH_{2n-1}$ , a group of the general structure



(Structure VI)

and a group of the general structure

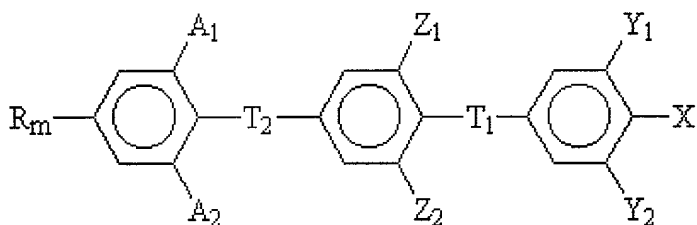


(Structure VII)

wherein  $R_x$  for both structures is selected from a group consisting of an alkyl group

- 5 having the general formula  $C_xH_{2x+1}$ , an alkenyl group having the general formula  $C_xH_{2x-1}$ , an alkoxy group having the general formula  $-OC_xH_{2x+1}$ , and an alkenoxy group having the general formula  $-OC_xH_{2x-1}$ .

- 10 16. A eutectic mixture of liquid crystal compounds comprising at least two liquid crystal compounds including at least one compound having the general structure



(Structure V)

wherein X is selected from the group consisting of F (fluoro), CN (cyano),  $OCF_3$  (trifluoromethoxy), and NSC (isothiocyanate);

- 15  $T_1$  is selected from the group consisting of a triple and a double covalent bond between two carbons;

$T_2$  is selected from the group consisting of a triple and a double covalent bond between two carbons; and,

$T_1$  is not equal to  $T_2$  when  $T_1$  or  $T_2$  is a double bond;

$Y_1$  and  $Y_2$  are a pair of substituents selected from the group consisting of H and F and  $Y_1$

5 =  $Y_2$ ;

$Z_1$  and  $Z_2$  are a pair of substituents selected from the group consisting of H and F and  $Z_1$

=  $Z_2$ ;

$A_1$  and  $A_2$  are a pair of substituents selected from the group consisting of H and F and  $A_1$

=  $A_2$ ;

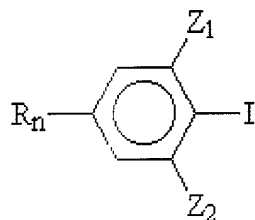
10 at least one of the pairs  $Y_1$  and  $Y_2$ ,  $Z_1$  and  $Z_2$ , and  $A_1$  and  $A_2$  is substituted with F; and,

$R_m$  is selected from the group consisting of selected from a group consisting of an alkyl group having the general formula  $C_mH_{2m+1}$ , an alkenyl group having the general formula  $C_mH_{2m-1}$ , an alkoxy group having the general formula  $OC_mH_{2m+1}$ , and an alkenoxy group having the general formula  $OC_mH_{2m-1}$ .

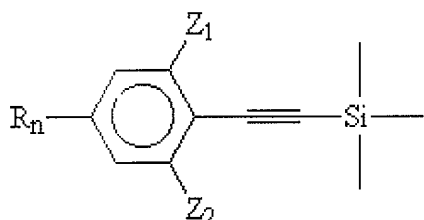
15

17. A method for preparing liquid crystal compounds, comprising the steps of:

- a) reacting an iodobenzene as shown in Structure 1 with trimethylsilyl acetylene in the presence of a catalyst and an amine to produce an trimethylsilylacetyl derivative as shown in structure 2;



(Structure 1)

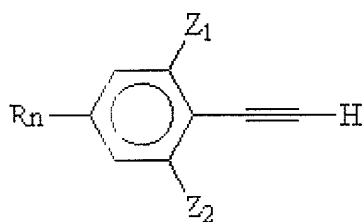


(Structure 2)

5 ;

b) isolating the trimethylsilylacetyl derivative shown in structure 2 from the reaction of the iodobenzene shown in structure 1 and trimethylsilylacetylene in the presence of the catalyst and the amine;

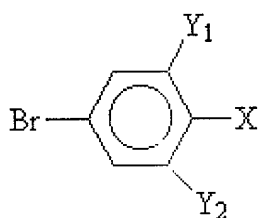
c) reacting the trimethylsilylacetyl derivative shown in structure 2 with a base to remove trimethyl silane and to give an unsubstituted product as shown in structure 3;



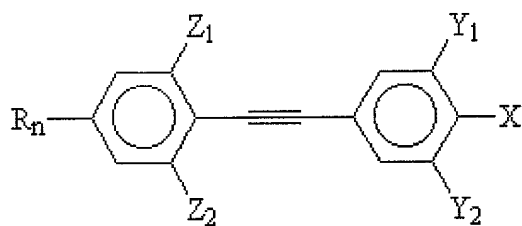
(Structure 3)

d) isolating the unsubstituted product as shown in structure 3 from the reaction of the trimethylsilylacetyl derivative shown in structure 2 with the base;

e) reacting the unsubstituted product as shown in structure 3 with a brominated, substituted benzene as shown in structure 3a to give a tolane product as shown in structure 4;



(Structure 3a)



(Structure 4)

f) isolating the tolane product shown in structure 4 from the reaction of the unsubstituted product as shown in structure 3 with the brominated, substituted benzene brominated, substituted benzene shown in structure 3a;

wherein X is selected from the group consisting of F (fluoro), CN (cyano), OCF<sub>3</sub>(trifluoromethoxy), and NSC(isothiocyanate);

$T_1$  is a triple bond;

$Y_1$  and  $Y_2$  are a pair of substituents selected from the group consisting of H and F, and  $Y_1 = Y_2$ ;

$Z_1$  and  $Z_2$  are a pair of substituents selected from the group consisting of H and F, and  $Z_1$

5  $= Z_2$ ; and,

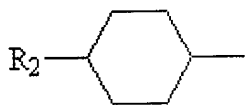
At least one of the pairs  $Y_1$  and  $Y_2$  and  $Z_1$  and  $Z_2$  is substituted with F;

$R_n$  is selected from the group consisting of an alkyl group having the general formula

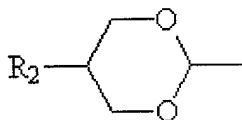
$C_nH_{2n+1}$ , an alkenyl group having the general formula  $C_nH_{2n-1}$ , an alkoxy group having

the general formula  $OC_nH_{2n+1}$ , an alkenoxy group having the general formula  $OC_nH_{2n-1}$ , a

10 group of the general structure



and a group of the general structure



wherein  $R_x$  for both structures is selected from a group consisting of an alkyl group

15 having the general formula  $C_xH_{2x+1}$ , an alkenyl group having the general formula  $C_xH_{2x-1}$ ,

an alkoxy group having the general formula  $OC_xH_{2x+1}$ , and an alkenoxy group having the

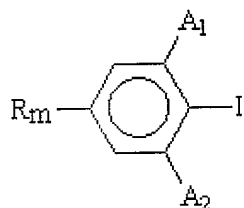
general formula  $OC_xH_{2x-1}$ .

18. The method for preparing liquid crystal compounds as set forth in claim 17, wherein the catalyst in steps a) and e) is  $\text{Pd}(\text{Ph}_3)_2\text{Cl}_2/\text{CuI}$ .

5 19. The method for preparing liquid crystal compounds as set forth in claim 17, wherein the amine in steps a) and e) is triethylamine.

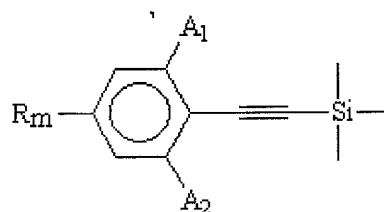
20. The method for preparing liquid crystal compounds as set forth in claim 17, wherein the base in step c) is  $\text{NaOH}$ .

10 21. A method for preparing liquid crystal compounds, comprising the steps of:  
a) reacting an iodobenzene as shown in Structure 5 with trimethylsilyl acetylene in the presence of a catalyst and an amine to produce an trimethylsilylacetyl derivative as shown in structure 6;



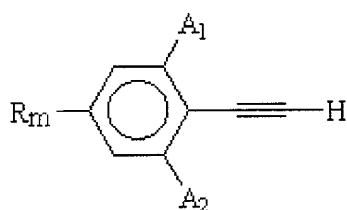
15 (Structure 5)





(Structure 6)

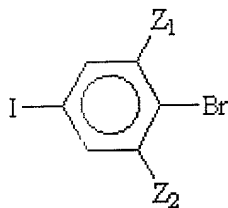
- 5 b) isolating the trimethylsilylacetyl derivative shown in structure 6 from the reaction of the iodobenzene shown in structure 5 and trimethylsilylacetylene in the present of the catalyst and the amine;
- c) reacting the trimethylsilylacetyl derivative shown in structure 6 with a base to remove trimethyl silane and to give an unsubstituted product as shown in structure 7;



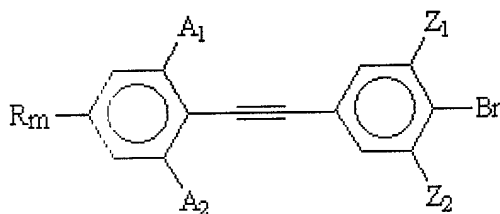
(Structure 7)

- 10 d) isolating the unsubstituted product as shown in structure 7 from the reaction of the trimethylsilylacetyl derivative shown in structure 6 with the base;
- e) reacting the unsubstuted product as shown in structure 7 with a substituted
- 15 bromiodobenzene as shown in structure 7a in the presence of a catalyst, an

amine, and triphenylphosphine to prepare a brominated, substituted tolane product shown in structure 8;



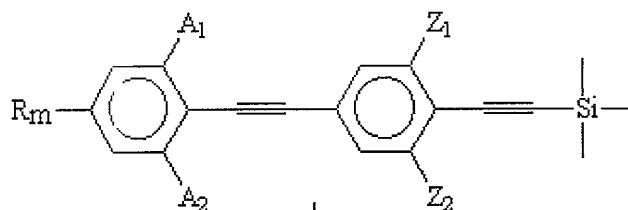
(Structure 7a)



(Structure 8)

f) isolating the brominated, substituted tolane product shown in structure 8 from the reaction of the unsubstituted product as shown in structure 7 with the substituted bromiodobenzene as shown in structure 7a in the presence of the catalyst, the amine, and triphenylphosphine;

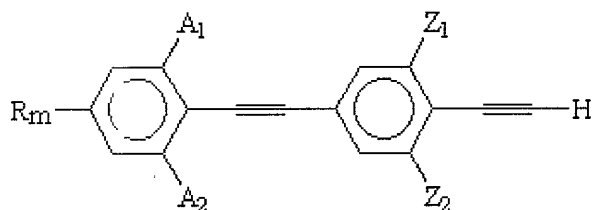
g) reacting the brominated, substituted tolane product shown in structure 8 with trimethylsilylacetylene in the presence of the catalyst, the amine, and triphenylphosphine to produce a trimethylacetyl derivative as shown in structure 9;



(Structure 9)

h) isolating the trimethylacetyl derivative shown in structure 9 from the reaction of the brominated, substituted tolane product shown in structure 8 with trimethylsilylacetylene in the presence of the catalyst, the amine, and triphenylphosphine;

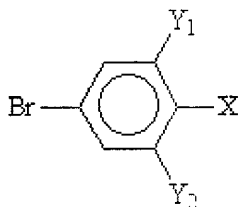
i) reacting the trimethylsilylacetyl derivative shown in structure 9 with a base to remove trimethylsilane and produce an unsubstituted product as shown in structure 10;



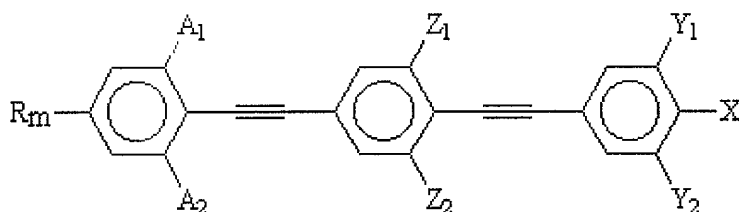
(Structure 10)

j) isolating the unsubstituted product shown in structure 10 from the reaction of the trimethylsilyl derivative shown in structure 9 with the base;

k) reacting the unsubstituted product shown in structure 10 with a substituted bromobenzene as shown in structure 10a in the presence of a catalyst, an amine, and triphenylphosphine to produce a bis-tolane product as shown in structure 11;



(Structure 10a)



(Structure 11)

5

l) isolating the bis-tolane product shown in structure 11 from the reaction of the unsubstituted product shown in structure 10 with the substituted bromobenzene shown in structure 10a in the presence of the catalyst, the amine, and triphenylphosphine;

10 wherein for the structures shown, X is selected from the group consisting of F (fluoro), CN (cyano), OCF<sub>3</sub>(trifluoromethoxy), and NSC(isothiocyanate);

T<sub>1</sub> is selected from the group consisting of a triple and a double covalent bond between two carbons;

15 T<sub>2</sub> is selected from the group consisting of a triple and a double covalent bond between two carbons; and,

T<sub>1</sub> is not equal to T<sub>2</sub> when T<sub>1</sub> or T<sub>2</sub> is a double bond;

$Y_1$  and  $Y_2$  are a pair of substituents selected from the group consisting of H and F and  $Y_1 = Y_2$ ;

$Z_1$  and  $Z_2$  are a pair of substituents selected from the group consisting of H and F and  $Z_1 = Z_2$ ;

5  $A_1$  and  $A_2$  are a pair of substituents selected from the group consisting of H and F and  $A_1 = A_2$ ;

at least one of the pairs  $Y_1$  and  $Y_2$ ,  $Z_1$  and  $Z_2$ , and  $A_1$  and  $A_2$  is substituted with F; and,

$R_m$  is selected from the group consisting of selected from a group consisting of an alkyl group having the general formula  $C_mH_{2m+1}$ , an alkenyl group having the general formula

10  $C_mH_{2m-1}$ , an alkoxy group having the general formula  $OC_mH_{2m+1}$ , and an alkenoxy group having the general formula  $OC_mH_{2m-1}$ .

22. The method for preparing liquid crystal compounds as set forth in claim **21**, wherein the catalyst in steps a), e), g), and k) is  $Pd(Ph_3)_2Cl_2/CuI$ .

15

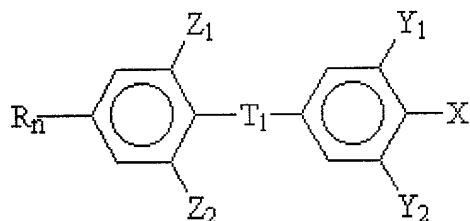
23. The method for preparing liquid crystal compounds as set forth in claim **21**, wherein the amine in steps a), e), g), and k) is triethylamine.

24. The method for preparing liquid crystal compounds as set forth in claim **21**, wherein

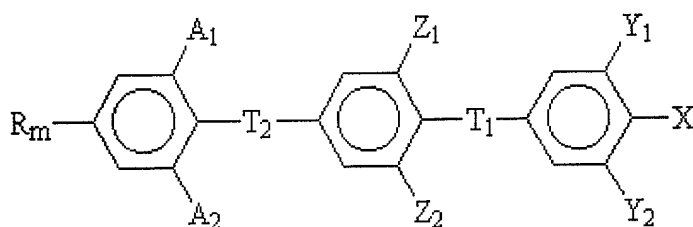
20 the base in steps c) and i) is NaOH.

## ABSTRACT

A new class of liquid crystal compounds is based on tolane and bis-tolane structures:



(Structure IV)



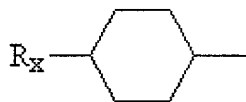
(Structure V)

in which  $X$  is a polar group such as  $F$  (fluoro),  $CN$  (cyano),  $OCF_3$  (trifluoromethoxy), or  $NCS$  (isothiocyanate) at least one of the pairs of sites  $Y_1$  and  $Y_2$ ,  $Z_1$  and  $Z_2$ , and for the bis-tolane derivatives,  $A_1$  and  $A_2$  are fluoro groups.

- 10  $T_1$  for the tolane derivatives is always a triple bond. For the bis-tolane derivatives,  $T_1$  and  $T_2$  are either both triple bonds or one of the two groups is a double bond with and the other remains a triple bond.

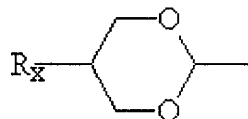
$R_n$  or  $R_m$  may be an alkyl group having the general formula  $C_nH_{2n+1}$ , an alkenyl group having the general formula  $C_nH_{2n-1}$ , an alkoxy group having the general formula

- 15  $OC_nH_{2n+1}$ , or an alkenoxy group having the general formula  $-OC_nH_{2n-1}$ . Additionally, for the tolane compounds,  $R_n$  may be a cyclohexyl substituent:



(Structure VI)

or a dioxane substituent:



5 (Structure VII)

in which  $R_x$  is an alkyl group having the general formula  $C_xH_{2x+1}$ , an alkenyl group having the general formula  $C_xH_{2x-1}$ , an alkoxy group having the general formula  $OC_xH_{2x+1}$ , or an alkenoxy group having the general formula  $OC_xH_{2x-1}$ .

These compounds exhibit useful nematic ranges and melting points. Also disclosed are

10 eutectic mixtures including these compounds.

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<b>DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION (37 CFR 1.63)</b>	<b>Attorney Docket Number</b> HRL033B	
	<b>First Named Inventor</b> Shin-Tson Wu	
	<b>COMPLETE IF KNOWN</b>	
	<b>Application Number</b>	/
	<b>Filing Date</b>	
	<b>Group Art Unit</b>	
<input checked="" type="checkbox"/> Declaration Submitted with Initial Filing	<b>OR</b>	<input type="checkbox"/> Declaration Submitted after Initial Filing (surcharge (37 CFR 1.16 (e)) required)
		<b>Examiner Name</b>

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

Polar Tolane Liquid Crystals

the specification of which (Title of the Invention)

☒ is attached hereto  
OR

☐ was filed on (MM/DD/YYYY) as United States Application Number or PCT International

Application Number and was amended on (MM/DD/YYYY) (if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56

I hereby claim foreign priority benefits under 35 U.S.C. 119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT international application having a filing date before that of the application on which priority is claimed.

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			<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
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I hereby claim the benefit under 35 U.S.C. 119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)	<input type="checkbox"/> Additional provisional application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto.

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U.S. Parent Application or PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)

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☒ Registered practitioner(s) name/registration number listed below

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Name	Registration Number	Name	Registration Number
Cary Tope-McKay	41,350		

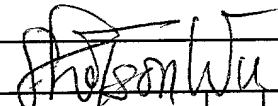
☐ Additional registered practitioner(s) named on supplemental Registered Practitioner Information sheet PTO/SB/02C attached hereto.

Direct all correspondence to: ☐ Customer Number or Bar Code Label

OR ☒ Correspondence address below

Name	Cary Tope-McKay				
Address	23852 Pacific Coast Highway #311				
Address					
City	Malibu	State	CA	ZIP	90265
Country	USA	Telephone	(310) 589-8158	Fax	(310) 943-2736

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor:		<input type="checkbox"/> A petition has been filed for this unsigned inventor			
Given Name (first and middle [if any])		Family Name or Surname			
Shin-Tson		Wu			
Inventor's Signature				Date	8/30/00
Residence: City	Northridge	State	CA	Country	USA
				Citizenship	USA
Post Office Address	19138 Frankfort St.				
Post Office Address					
City	Northridge	State	CA	ZIP	91324
				Country	USA

☒ Additional inventors are being named on the 2 supplemental Additional Inventor(s) sheet(s) PTO/SB/02A attached hereto

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<b>DECLARATION</b>	<b>ADDITIONAL INVENTOR(S)</b> <b>Supplemental Sheet</b> Page <u>1</u> of <u>2</u>
--------------------	---

<b>Name of Additional Joint Inventor, if any:</b>		<input type="checkbox"/> A petition has been filed for this unsigned inventor			
Given Name (first and middle [if any])		Family Name or Surname			
Zheng		Chai			
Inventor's Signature	Zheng Chai			Date	9/30/00
Residence: City	Richardson	State	TX	Country	USA
				Citizenship	P.R. China
Post Office Address					
430 Buckingham Rd. #221					
Post Office Address					
City	Richardson	State	TX	ZIP	75081
				Country	USA
<b>Name of Additional Joint Inventor, if any:</b>		<input type="checkbox"/> A petition has been filed for this unsigned inventor			
Given Name (first and middle [if any])		Family Name or Surname			
Inventor's Signature				Date	
Residence: City		State		Country	
				Citizenship	
Post Office Address					
Post Office Address					
City		State		ZIP	
				Country	
<b>Name of Additional Joint Inventor, if any:</b>		<input type="checkbox"/> A petition has been filed for this unsigned inventor			
Given Name (first and middle [if any])		Family Name or Surname			
Inventor's Signature				Date	
Residence: City		State		Country	
				Citizenship	
Post Office Address					
Post Office Address					
City		State		ZIP	
				Country	

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## DECLARATION

ADDITIONAL INVENTOR(S)  
Supplemental Sheet  
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<b>Name of Additional Joint Inventor, if any:</b>		<input type="checkbox"/> A petition has been filed for this unsigned inventor			
Given Name (first and middle [if any])		Family Name or Surname			
Larry R.		Dalton			
Inventor's Signature	<i>Larry R. Dalton</i>			Date	8/29/2000
Residence: City	Silverdale	State	WA	Country	USA
Post Office Address	10965 Warren Rd. NW				
Post Office Address					
City	Silverdale	State	WA	ZIP	98383
				Country	USA
<b>Name of Additional Joint Inventor, if any:</b>		<input type="checkbox"/> A petition has been filed for this unsigned inventor			
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